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Propulsion Report 181

A SIMULATOR FOR A HYDROCARBON
RAMROCKET FUEL GAS GENERATOR -
FIRST PHASE DEVELOPMENT

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*Exchange scientist from United States Air Force, Aero Propulsion and Power Laboratory

SUMMARY

Development was undertaken of a combustor to simulate the effluent of a solid hydrocarbon propellant ramrocket fuel gas generator, for use as a research tool. In this first phase of the work, the combustor was limited to an operating pressure of 350 kPa and was fuelled by a mixture of toluene, ethylene and oxygen. The exhaust effluent was analysed for the compositions of both the gaseous and solid phases, and the results were compared with both theoretical predictions and published data from tests with a solid propellant gas generator. The work provides a basis for further development of simulator technology.



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POSTAL ADDRESS: Director, Aeronautical Research Laboratory,
P.O. Box 4331, Melbourne, Victoria, 3001, Australia

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1. INTRODUCTION

The development of ramrocket or ducted rocket propulsion systems requires research in many component areas. Two general subsets of these are the gas generator, the effluent of which functions as a fuel source, and the ramjet combustor or ramburner, in which the fuel from the gas generator is burned with incoming air. The material burned in the gas generator is essentially a solid propellant, similar to solid fuels used for rocket propulsion, but with a much lower proportion of oxidant. The fuel-rich effluent from the gas generator is injected into the ramburner where it is mixed and burned with the air which enters through the external inlets of the missile.

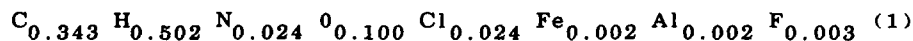
A device which simulates the gas generator, burning a mixture of readily available gaseous and/or liquid fuels, would be a most valuable tool for ramrocket research. For example, the effects of additives or changes in fuel composition on ramburner operation could be evaluated with a simulator, thus decreasing the time and costs associated with manufacturing and handling new solid propellant fuel grains for each evaluation. In some cases, a simulator may actually be better suited to investigation of the effects of individual fuel constituents than is a solid propellant gas generator. It may also be practical to use a simulator for the development of mechanical components of the gas generator such as variable area nozzles, actuators and control systems.

In ramburner research, basic investigation of such aspects as combustor flow fields would be much easier to perform without the added complication of firing a solid propellant gas generator nearby. A great deal of ramburner testing could also be accomplished to determine combustion efficiencies, pressure losses, high temperature materials performance, and to evaluate various components such as inlet covers and nozzles. Several advantages are apparent here: among these are direct fuel flow control, ease of varying the fuel formulation, lower cost, and relative safety. Laboratories which are excluded from experimental ramrocket research due to their proximity to inhabited areas, or due to other restrictions associated with the use of solid propellants, would profit from the availability of a practical simulator.

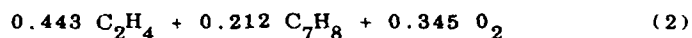
In the past it has been common practice for ramrocket combustor research to be carried out using fuels which provide quite loose simulation of a gas generator efflux, including unheated gaseous or even liquid hydrocarbons¹. In this report, a device is described which was designed to burn a mixture of fluids producing an efflux closely simulating that of a solid hydrocarbon gas generator, in terms of temperature and chemical composition. While this initial version of the simulator operated at low pressures relative to typical solid gas generators, it served to demonstrate the feasibility of the approach and to lay the groundwork for continued development of the concept.

2. FUEL COMPOSITION

The solid gas generator fuel chosen for simulation is a hydrocarbon propellant of current interest^{2,3}, consisting of a mixture of polystyrene, ammonium perchlorate, polybutadiene binder, and small amounts of other compounds such as combustion catalysts. A significant benefit in selecting this particular fuel is that a report published by Zaccardi and McGregor⁴ provides more information about its exhaust gas composition than is available for any other relevant fuel. While it is not practical to simulate each constituent of the propellant or its function on an individual basis, it is possible to represent the fuel as if it were a compound comprised of the elements from each component in the mixture, thus obtaining a guide to the overall elemental composition required in a reasonable combination of simulator feed materials. Reducing the solid propellant formulation in this way yields overall atom fractions as follows:



Notwithstanding the importance of the minor constituents to both primary and secondary combustion processes with the solid propellants, in the interests of simplicity only carbon, hydrogen and oxygen were included in the experimental simulator fuel. The omission of the other elements in Formula (1) was thought to be an issue of minor importance in development of the simulator concept, and in applying it to many areas of ramburner research. The approach was to formulate a mixture of ethylene, toluene and oxygen such that the relative proportions of carbon, hydrogen and oxygen in the mixture were representative of those in the solid propellant. Precise replication of the relative atom fractions of these elements in Formula (1) may be achieved using the mixture:



Ethylene was chosen because of its relatively high vapour pressure, making external pumping unnecessary and, since it could be injected in gaseous form, it minimised problems with atomisation, evaporation and mixing. Toluene was chosen because of its relatively high carbon/hydrogen ratio. A gaseous fuel would have been preferred; however, the only one with an adequate carbon/hydrogen ratio is acetylene, which is unsafe for use at the pressure levels of interest. Benzene was avoided due to toxicity considerations, but remains a possible candidate. Noteworthy features of the fuels chosen for the simulator are that all are readily available, they require no processing before use, and they are relatively safe to handle. The safety of a simulator using such a mixture of fuels, compared with a solid propellant gas generator, is further enhanced by maintaining adequate separation between the fuels and oxidiser until their controlled injection into the simulator.

The products of combustion of the above fuel mixtures, as predicted by thermochemical equilibrium theory, may be calculated using an existing NASA computer code⁵. Results of such calculations are shown in Table 1 for the solid gas generator propellant and the fuel formulation in Formula (2), both burning at a pressure of 7000 kPa. Neglecting the products formed from the additional minor constituents in the solid propellant, there is a broad similarity between the two sets of results. There are, however, notable differences, associated with the temperatures attained at equilibrium, which arise from the different heats of formation of the initial reactants in the two fuels. These are manifested largely in the different amounts of carbon monoxide, methane and hydrogen in the exhaust efflux.

Table 1 illustrates only one reason why it would be unrealistic to expect precise simulation to be achieved directly by the above approach. An important purpose of the exercise was therefore to gain an appreciation of the differences occurring in practice between the efflux properties of the solid propellant and the simple simulator fuel, with a view eventually to developing methods for compensating for the differences. These methods may include adjustment of the simulator fuel formulation, direct addition of material to the simulator exhaust, or even selective extraction of gaseous or solid material.

3. EXPERIMENTAL APPROACH

For experimental convenience, the combustor pressure in this initial phase was limited to 350 kPa. The experiments were designed to:

- (a) Identify the requirements for further development in terms of overall combustor geometry, fuel formulation, higher pressure operation, analysis methods, and equipment needed.
- (b) Provide results for comparison with theoretical predictions and the results of Zaccardi and McGregor⁴, and thus gain insight into the combustion processes in both the simulator and the solid gas generator.

The next phase in this approach could involve a simulator designed to operate at high pressure, say 3500 kPa. While an efflux having appropriate composition can be obtained at low pressures, this higher value would ensure a choked nozzle at the simulator outlet over a meaningful ramrocket flight envelope, and a level of total pressure in the efflux which is more representative of typical solid propellant gas generators. This would provide a most useful experimental tool while still operating at reasonable working pressures in terms of fuel sources and safety requirements.

4. EXPERIMENTAL APPARATUS

4.1 Combustor

Once the appropriate mixture of fuels was determined, designing the combustor involved taking advantage of the ease of mixing and burning ethylene with oxygen to form a high temperature flame which aided in the evaporation and reaction of the toluene. The choice of combustor configuration was based on a balance between internal gas velocities, residence time, heat loss, and practical size in terms of fabrication, installation, and instrumentation.

As shown in Figure 1, the combustor was fabricated from a cylindrical stainless steel tube, 50 mm in diameter and 300 mm long, held between flanges by four compression springs. This method of construction was chosen for relative ease of modification, to allow for thermal expansion and to provide relief of any unexpected over-pressure. Gaseous ethylene entered through the stainless steel top

closure via an internal manifold consisting of 12 holes, 1.5 mm in diameter, oriented at 45° to the vertical. The ethylene mixed with half the total oxygen flow, injected through a similar manifold with 12 holes, 1.7 mm in diameter, to form a relatively hot primary combustion zone around a pressure-atomising spray nozzle for liquid toluene injection. The remaining oxygen was injected radially through 12 holes, also 1.7 mm in diameter, distributed around the circumference of the combustor tube 75 mm from the top. Dividing the oxygen injection between two separate locations in this way kept the primary zone flame temperature to a manageable level. A standard automotive spark plug, mounted 22 mm from the top of the combustor, was used for ignition. Combustor pressure was measured with a single transducer connected at the midpoint of the 50 mm diameter tube, while combustor temperature was measured immediately upstream of the entrance to the exhaust nozzle.

The stainless steel exhaust nozzle consisted of an orifice with a 9.22 mm throat, following a 14.2 mm diameter cylindrical entry section approximately 81 mm long. Four holes, 2 mm in diameter, were equally spaced around this entry section at a point 29 mm from the nozzle exit plane to provide access for obtaining exhaust gas samples. This design was chosen to duplicate, as far as possible, the internal contour and gas sampling locations of the converging-diverging nozzle described in the Zaccardi and McGregor report⁴, to minimise possible sources of difference when comparing results. The combustor was oriented to fire vertically downwards to ensure that unburned liquid fuels were easily cleared.

4.2 Fuel and Oxygen Supply

The required flow rates of fuels and oxygen to produce 350 kPa in the simulator were initially estimated using thermochemical equilibrium theory to calculate idealised values for the temperature, molecular weight, and ratio of specific heats of the combustion products, and by assuming isentropic flow of a perfect gas through the choked nozzle at the simulator exit.

Actual flow rates were determined after sufficient operating experience was obtained to develop a proportional relationship between pressure and flow rate which, in effect, lumped the unknown nozzle discharge coefficient, deviations from perfect gas properties and the molecular weight of the exhaust together into a single constant.

The supply system for the fuels, oxygen, and nitrogen is shown schematically in Figure 2, along with appropriate instrumentation for flow measurement. At the top of the Figure, oxygen is shown as supplied from a standard G-size cylinder, connected to a manually adjustable pressure regulator. A manual control valve was included for fine adjustment of flow rate. The oxygen mass flow was measured with two separate choked orifices, 1.25 mm in diameter. The use of two separate orifice lines facilitated the selection of either half or full oxygen flow during start-up and shut-down. Remotely actuated pneumatic ball valves were used for on-off control of each line. Following a check valve, the oxygen flow was divided between the upper and lower manifolds of the combustor.

The ethylene system was of similar design and construction, using a single 1.7 mm orifice for flow rate measurement. All orifices used for flow measurement were designed using thick orifice criteria for critical flow as described in Reference 6. However, due to their small size and consequent susceptibility to errors in fabrication, each choked orifice was individually calibrated against a standard orifice plate. Their measured discharge coefficients ranged from 0.88 to 0.92.

The design operating pressure for both gases was 1380 kPa upstream of the choked orifices to provide the desired flow rate for the 350 kPa combustor pressure.

The toluene source was a pressurised tank, charged to 2000 kPa by gaseous nitrogen. The flow rate of nitrogen into the tank was restricted by an orifice 0.25 mm in diameter so that, in the event of an emergency, the pressure over the toluene could be dumped through a valve with a much larger effective throat size. The toluene flow to the combustor was measured as described in Section 5. A remotely actuated ball valve was used for on-off control, with a check valve installed immediately before the combustor to prevent reverse flow.

Nitrogen was used to purge the fuel and oxygen lines via the combustor and to clear the gas sampling system. The flow rate of nitrogen from its 2000 kPa source was fixed by a 2mm diameter orifice. Again, check valves were installed to prevent reverse flow and inadvertent mixing of the fuels prior to entering the combustor.

4.3 Exhaust Ducting

An enclosed exhaust duct was installed, as shown in Figure 3, consisting of 50 mm pipe leading to a horizontally oriented ejector fabricated from a 150 mm diameter stainless steel tube, 775 mm long, in which the exhaust products were mixed with ambient air and burned off. The exhaust duct terminated 70 mm inside the entrance to the ejector, in a small nozzle. The resultant back-pressure in the exhaust duct made it easier to obtain samples of the efflux downstream of the combustor exit nozzle when it was necessary to do so. A spark plug, installed at the entrance to the ejector, had electrodes reaching to the central exhaust jet to ensure that the effluent/air mixture ignited.

5. RIG INSTRUMENTATION

Instrumentation on the test rig was limited to that required for measurement of fuel and oxygen flow rates together with combustion parameters. Seven channels of data were electronically recorded during simulator combustion tests. Pressure transducers were used for the oxygen, ethylene, and combustor pressures. The oxygen and ethylene transducers had a 3500 kPa range, chosen for the high pressure conditions which occurred when their respective downstream ball valves were closed. The combustor pressure was measured with a 1750 kPa transducer. All temperatures were measured with chromel-alumel (K-type) thermocouples. Oxygen and ethylene temperatures were measured with 1 mm diameter thermocouples for quick response, while a more robust 2 mm diameter thermocouple was chosen for combustor temperature measurements. The toluene flow rate was measured with a Micro Motion flowmeter having an electrical output directly proportional to mass flow rate, thereby eliminating the need for the specific gravity corrections commonly required of volume flow measurements. The seven channels of data were recorded with a data logger. Analogue gauges were used for direct indication of pressure in the nitrogen system, the combustor, the exhaust duct and the exhaust gas sample cylinder.

6. TEST PROCEDURE

Before testing, the ethylene and oxygen pressure regulators were pre-set while flowing the corresponding gas. Immediately prior to lighting the combustor, it was purged with nitrogen for 5 to 10 seconds. Then the ignition coils were switched

on to both the combustor and the exhaust ejector, along with a single momentary switch which turned on the ethylene and half the total oxygen flow (only one oxygen valve open). Ignition was made much easier by having the oxygen flow lead the ethylene by approximately one second. This was accomplished with a small restriction installed in the pneumatic control line which opened the ethylene valve. After ignition, which normally followed a one to two second delay for valve actuation, the ignition switch was released and the remaining momentary switch was pressed to start the toluene flow and open the second oxygen valve. After the combustor flow stabilised, the gas sampling valve was remotely actuated for the specified time, depending on the type of exhaust sampling being undertaken. During the entire test, the exhaust gases were burned off at the ejector in an intense yellow-white flame one to two metres long. Figure 4 shows a photograph of the combustor in operation.

Shutdown was normally accomplished by first releasing the toluene/oxygen switch, then the ethylene/oxygen switch two to three seconds later. This procedure ensured that any residual liquid toluene was burned off by the ethylene/oxygen flame. A short nitrogen purge followed. During the entire test, fuel flow and combustor data were recorded for subsequent reduction. The only transient characteristics noted were associated with the hardware heating up from the initial time of ignition.

7. EXHAUST SAMPLING AND ANALYSIS

An important and exacting aspect of this work was the quantitative assessment of the results relative to the goal of accurately simulating a solid propellant gas generator. Measurements of combustor temperature and pressure, while reassuring if in reasonable agreement with expectation, are not enough. Only a thorough analysis of the exhaust products, both solid and gaseous, can reveal how well the combustor simulates a real gas generator and/or how closely it agrees with theoretical predictions.

A check on the validity of the exhaust analysis is available in the form of an audit of the elemental constituents found in the products of combustion. This can be carried out by comparing the balance of the total atomic content of the exhaust products with that known to exist in the fuel mixture.

7.1 Solid Products

Because a large proportion of the efflux was predicted to be solid carbon, it was important to account for the particulate products in the exhaust as well as gases. As will be seen later, measurement of the relative rates of production of solid and gaseous materials may be redundant if accurate chemical analyses of the two phases are available. Nonetheless, for experimental purposes, measurement of production rates as well as chemical analyses were attempted in this work. Two methods of capturing solid samples were investigated, referred to here as wet filtering and as dry filtering, using a common probe system for extracting a sample from the exhaust duct downstream of the combustor exit nozzle.

7.1.1 Probe System

In order to obtain a representative sample of the solid material expelled from the combustor, a sample probe was fabricated from 6 mm tubing and installed in the 50 mm exhaust duct with the probe entrance approximately 25 mm downstream of the combustor exit nozzle. As shown in Figure 3, the sample probe was connected to a pneumatically operated three-way valve so that an initial flow could be established through the tube before sampling. During the measurement period, the valve directed the sample to the particle collection equipment. No attempt was made at isokinetic sampling, the assumption being, at least initially, that the particles were small enough to follow the flow into and around the sample probe. Also, the distribution of particles across the exhaust duct was assumed to be uniform.

7.1.2 Wet Filtering

This technique used a flask containing approximately 60 ml of water, connected to the probe and valve system, to capture the solid particles as the exhaust sample bubbled through the water. Downstream of the water bath, the remaining gaseous sample was passed through a cooling coil to condense as much water vapour as possible, then to a filter vessel containing glass wool to remove residual moisture. Based on chemical equilibrium predictions, no water vapour was expected in the exhaust; thus, every effort was made to remove moisture from the sample downstream of the water bath. Finally, the gas flowed through a totalising gas volume meter. After the test, the particle-laden water was poured through a filter paper of known weight. The filter paper was dried for two hours at 120°C and weighed again to determine the amount of solid material present.

7.1.3 Dry Filtering

The use of a dry filtering technique has the potential benefit of eliminating ambiguities in the volume measurement due to water vapour introduced when the relatively hot exhaust gas is bubbled through the water-filled flask. After first trying a 36 mm diameter commercially-made gravimetric analysis filter, and finding it too small, a specially constructed assembly utilising a piece of laboratory filter paper mounted across a 110 mm diameter steel tube was used to collect the solid particles. This filter, having almost nine times the collection area of the commercial version, worked reasonably well. The sampling probe and gas volume measurement system were the same as in the wet filtering system.

In order to maximise the validity of comparison of the results from the two sampling techniques, the material collected on the dry filter was also "cooked" at elevated temperature to drive off volatile products which may have been present before weighing the sample. At this stage of the investigation pure carbon particles were the target of the solid sampling effort. In the light of subsequent analyses, discussed in Section 9 below, it would have been advantageous to have weighed the sample as collected, without further treatment.

7.2 Gaseous Products

7.2.1 Gas Sampling System

The arrangement used for exhaust gas sampling is shown in Figure 5. In order to be consistent with the experimental setup used by Zaccardi and McGregor⁴, the gas sample was taken from one of the side ports of the exhaust nozzle, just upstream of the choke point, as described in Section 4.1. The copper sample line, 6.3 mm in diameter, was closely coupled to a water-jacketed section to quench any on-going reactions as quickly as possible. The measured gas temperature downstream of the cooling section was approximately 120°C. As may be seen in Figure 5, the gas sampling line was set up to flow continuously to the ejector to ensure that, at the moment of sampling, the captured gases were representative of the mixture issuing from the combustor. A flow-restricting valve in this line provided pressure to aid in filling the sample container. When the air-operated ball valve was opened to obtain a sample, the gases passed through a 1.6 micron glass microfibre filter, 37 mm in diameter, which served two functions. Firstly, it prevented solid material from

entering the sample container, and secondly, it provided a means of capturing a solid sample simultaneously with the gas sample for subsequent analysis. The sample bottle was evacuated with a vacuum pump prior to the test, and filled with the sample to a pressure of approximately 100 kPa to facilitate injection into the gas chromatograph.

7.2.2 Gas Chromatography

Gas chromatography was chosen as the single most useful technique to analyse the exhaust samples for permanent gas composition. A Perkin Elmer Model F-11 chromatograph, with a thermal conductivity detector, was used for this purpose. While dated by modern standards, this instrument was found to be suitable for most of the required analyses and confirmed the value of chromatography in this research. Twelve different gases were identified and quantified in samples taken from the simulator, using the columns and operating conditions set out in Table 2. Water, benzene, and toluene calibrations were obtained by direct injection, in liquid form, of the amount required to get an appropriate fraction of vapour in the sample volume.

Analysis of the captured sample showed a smaller quantity of water, benzene, and toluene present than the calculated saturation levels of each at room temperature. Thus, it was considered to be unnecessary to heat the sample lines or the gas sampling valve on the chromatograph to ensure that these species were in gaseous form for the analysis. The sample loop volume on the gas sampling valve was 1 cm^3 for all gases except benzene and toluene vapour, for which a 5 cm^3 loop was used to obtain adequate sensitivity. Argon was used as the carrier gas for all the analyses.

8. OBSERVATIONS OF TEST HARDWARE PERFORMANCE

In general, the experimental hardware worked well as initially designed. The combustor functioned effectively, although the use of compression springs in the assembly proved to be unnecessary and could have been replaced by a standard flanged construction. Run times in excess of one minute were accomplished with no evidence of mechanical difficulty or combustor overheating, while measured combustion temperatures approximated the predicted values. The only routine maintenance required was an occasional disassembly to clean out accumulated soot

deposits. The 'O'-rings in the upper manifolds were replaced once after many test runs. No sign of leakage was detected. Trials of a cylindrical heat shield fitted to the brass bodied spray nozzle in the centre of the combustor actually proved to be counterproductive, as the shield apparently acted as a flameholder - the evidence being a green flame and a melted spray nozzle.

The fuel, oxygen and nitrogen systems were essentially adequate as installed, requiring only minor modification. As the fuel flow rates were increased to design levels during initial combustor checkout, it became necessary to install a heat exchanger in the ethylene line upstream of the flow measuring section to keep the gas temperature above 0°C for accurate temperature measurement. No other modifications were required. The only serious operational difficulties were two separate failures of check valves immediately ahead of the combustor, resulting in mixing ethylene and oxygen in a short length of tubing next to the combustor. The tubing immediately became red hot and, on one occasion, burned through. The normal shut-down procedure was adequate to prevent further damage. Although flow rate adjustments were tedious without remotely actuated control valves, the system was reliable and safely controlled. As described in Section 6, the ethylene and oxygen pressure regulators could be set only while flowing the corresponding gas. This feature, which usually required two people and wasted a moderate amount of gas each time, could be improved with remotely actuated pressure regulators and flow control valves. The use of choked orifices for gaseous flow rate measurements was adequate; however, commercially available flowmeters, providing greater range and accuracy, would be preferable.

Burning off the effluent after mixing the air in the ejector proved to be a safe, reliable method to dispose of the exhaust products. Some experimentation was required to determine the optimum ejector size. Although the configuration was not particularly critical, a smaller ejector used for operating with lower exhaust flow rates failed to provide adequate flame stabilisation at the design operating conditions. Plans to run the existing unit over the broad range of flow rates required for testing at different simulator pressures will probably require further adjustments.

The gas sampling system evolved from initial attempts at collecting solid samples for gravimetric analysis. Even though the sample container was approximately three metres from the simulator, the flow rate of sample gas was such that achieving a representative quantity was assured. Sensitivity of the sample to

variations in line pressure and quenching temperature was not determined. Also, the actual point of sampling on the side of the exhaust nozzle, chosen to be similar to that used by Zaccardi and McGregor⁴, was not confirmed as the optimum location to use on the simulator.

9. RESULTS AND DISCUSSION

9.1 Analysis of Zaccardi and McGregor's Results

The work of Zaccardi and McGregor⁴ provided the only measured exhaust effluent data from a solid hydrocarbon gas generator which is currently known to be available. Their reported data from a representative test firing is reproduced in Table 3, together with their predictions based on thermochemical equilibrium calculations. Only gaseous species are included; although they captured a significant amount of solid material (reckoned to be almost pure carbon), this was not quantified as a proportion of the total effluent. Their predicted analysis of the gaseous phase can be shown to be in close agreement with the left hand column in Table 1 with the carbon removed.

As discussed in Reference 4, significant differences were evident between the measured and predicted outputs. An important discovery was the amount of benzene present in the sample. This identified the need to look for heavy hydrocarbons in the exhaust products captured in the present experiments.

Based on the conclusion in Reference 4 that the solid phase output was almost pure carbon (especially when elemental constituents other than carbon, hydrogen and oxygen are neglected) the atomic ratio of oxygen to hydrogen in the exhaust gas analysis should match that in the fuel. O/H ratios are listed below for the fuel formulation represented by Formula (1), and for both the predicted and measured gas analysis data in Table 3.

SOURCE	O/H
Formula (1)	0.199
Table 3 predicted	0.193
Table 3 measured	0.274

The ratio from Formula (1) and the predicted figure are slightly different, probably because the input data for the calculations performed by Zaccardi and McGregor were not duplicated exactly by Formula (1). A much more significant discrepancy exists in the measured data, in the form of a deficiency of hydrogen relative to oxygen. Numerous factors could have contributed to the discrepancy, including:

- (a) emission of hydrogen in gaseous form (either free or attached to other elements) which was not registered accurately in the gas analysis.
- (b) inexact correspondence between the captured gas and solid samples in terms of the specific test firing, or the instant of sampling during the test firing; this aspect is not clear from Reference 4.
- (c) the presence of undetected hydrogen atoms in the solid phase product.

The last of these possible factors would be consistent with results from the present experiments which are discussed in Section 9.2 below. Zaccardi and McGregor's conclusions as to the composition of their captured solid material were based mainly on an experiment in which the solid sample was subjected to successively higher levels of heating whilst its loss of mass was monitored. A complete chemical analysis of the sample, which may have been more conclusive, was evidently not available.

Consideration of these issues may be helped by reference to the C-H-O ternary diagram shown in Figure 6. Assuming that the chemical processes in the gas generator involve only the three atomic species carbon, hydrogen and oxygen - an approximation in the case of Zaccardi and McGregor's solid propellant gas generator - then changes in the make-up of the three-component system which occur during the combustion process may be plotted on this graph. This procedure exploits the property of an equilateral triangle where the sum of the perpendiculars drawn from any point within the triangle to each of the three sides equals the length of the perpendicular drawn from a vertex of the triangle to the opposite side. The three coordinates of any point on the graph, defined by the prevailing atom fractions of the respective elemental species, thus always add up to unity. The point shown in Figure 6 as representing Zaccardi and McGregor's fuel is derived from Formula (1), by notionally eliminating the minor constituents and recalculating the C:H:O atom fractions as 0.36:0.53:0.11.

In a closed three-component system, when a single phase decomposes into two separate phases, the compositions of the original phase and the two product phases are "tied" by mass balance criteria such that, on the ternary diagram, the points representing the three phases must lie on a straight line (the tie line). Such a line appears on Figure 6 connecting the point representing the fuel with the two points representing the products of combustion predicted by thermochemical equilibrium theory. The solid phase is pure carbon (at the upper vertex of the triangle) and the gaseous phase has coordinates calculated from the first column of figures in Table 3, after neglecting the minor atomic constituents so that only carbon, hydrogen and oxygen are involved. These latter coordinates define a point which lies on a line which may be termed the carbon deposition boundary, below which solid carbon cannot exist in thermochemical equilibrium with a gas phase⁷.

Also shown in Figure 6, as a solid triangle, is a point representing the measured composition of Zaccardi and McGregor's gaseous efflux. This is calculated from the second column in Table 3, again after neglecting minor atomic species. It is now possible to draw a second tie line which represents the fuel decomposition which actually occurred, if the gaseous product phase is assumed to be accurately represented by the measured data in Table 3. A precise composition of the solid phase product which would correspond with this second tie line cannot be determined from the information available. If it is assumed to have contained only carbon and hydrogen (still neglecting the elemental constituents in minor fuel additives, for the purpose of this discussion), then it would be represented by the solid square shown on the C-H boundary. The C:H atom fraction at this point is 0.46:0.54, which translates to a C:H mass fraction of 0.91:0.09. In view of the fact that most if not all of the hydrogen associated with the solid phase would be tied to carbon, the mass fraction of pure carbon would be appreciably less than 0.91. The total solid:gas mass fraction in the efflux, calculated as set out in Appendix 1, would be 0.33:0.67.

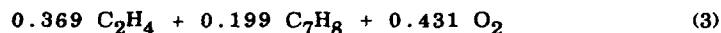
It is worth reiterating that the above conjecture on the nature of the solid material issuing from Zaccardi and McGregor's gas generator is based on the assumption that the compositions of the fuel and gaseous efflux in the experiment of Reference 4 are accurately represented by Formula (1) and the right hand column of Table 3 respectively. It is more than conceivable that a number of factors contributed to the discrepancy in the O:H ratio discussed earlier, and that the tie line representing the actual reaction lay somewhere between the two extremes shown on Figure 6. However, it seems safe to conclude that:

- (a) the actual combustion process did not reach anywhere near thermochemical equilibrium.
- (b) the solid phase product was not pure carbon, but more likely a "soot" containing a complex mixture of carbon, bound hydrogen and some adsorbed hydrocarbons.

9.2 Experimental Simulator Results

9.2.1 Fuel Composition

Much of the experimental program was conducted prior to accurate information on the solid propellant formulation becoming available. For this reason the results discussed in the following Sections were obtained with a simulator fuel mixture which was different from that detailed in Formula (2). The actual mixture was :



This was obtained with flow rates to the combustor of:

C_2H_4	0.272 kg/min
C_7H_8	0.483 kg/min
O_2	0.363 kg/min

This experimental mixture was somewhat less fuel-rich than that of Formula (2) and the solid propellant, so that the following results are only indirectly comparable with those of Reference 4.

9.2.2 Gas Phase Exhaust Analysis

Exhaust gas samples were captured and analysed as described in Section 7.2 above. Table 4 shows representative results of such an analysis, along with corresponding thermochemical equilibrium predictions. It is obvious that, like Zaccardi and McGregor's data, significant differences exist between the measured and predicted exhaust gas compositions. The data indicates that a greater proportion of hydrogen was emitted in unburned hydrocarbons than is predicted by equilibrium

calculations. This would also have limited the amount of carbon emitted in solid form from the simulator. As noted above, direct comparisons between the simulator results shown in Table 4 and Zaccardi and McGregor's measurements in Table 3 are difficult to make because of the relatively oxygen-rich flow rates used in the simulator.

9.2.3 Solid Phase Exhaust Analysis

Section 7.1 above outlines the two methods tried for direct measurement of the amount of solid matter in the efflux of the experimental simulator. In both cases the captured samples were subjected to washing and/or heat treatment prior to weighing, processes which may have caused loss of material which could not be accounted for. It should also be noted that the different exhaust sampling operations, including the gas sampling discussed in the previous Section, were conducted at different times, and matching of the respective results is therefore subject to experimental error arising from day to day variations in rig conditions, particularly the preset fuel and oxygen flow rates.

The measurement which was made of the mass of the particulate material yielded by the wet filtering process could be interpreted two ways:

- (a) If the material was assumed to be pure carbon, then knowledge of the corresponding volume of captured gaseous products led directly to a solid:gas mole fraction of 0.10:0.90.
- (b) If the captured gas was assumed to have the analysis in Table 4, then the solid:gas mass fraction could be calculated to be 0.06:0.94.

The wet filtering technique appeared to capture most of the solid matter arriving at the water bath, but uncertainties in the gas volume measurement due to unknown water vapour content rendered the calculated mole and mass fractions suspect. A mass flow measurement of the sample before it reached the water could solve this problem; however, doing this accurately enough with a mix of solid particles and combustion gases of uncertain molecular weight would present a complex problem in itself. Gas volume corrections for water vapour content at the minimum temperature of the cooling coils may also have helped.

The dry filtering technique yielded solid:gas ratios approximately double those quoted above. These results are arguably more reliable, but remain subject to the experimental uncertainties discussed earlier. Clearly, the quantitative solid sampling techniques require more development before the results can be applied with any confidence.

As described in Section 7.2.1 above, solid material was also filtered from the gaseous efflux at the inlet of the sample container during gas sampling operations. Inspection of some of the material trapped in the filter assembly revealed a damp consistency and an odour which suggested unburned hydrocarbons. This is perhaps not surprising, since a major component of the simulator fuel was liquid toluene, possibly making the simulator more prone to this occurrence than the solid propellant gas generator. A sample of the material, captured during the test which yielded the results shown in Table 4, was commercially analysed without further treatment, with the results shown in Table 5. These figures show that the amount of hydrogen trapped in the solid matter was significant, especially when expressed in terms of atom fractions.

9.2.4 Balance of Exhaust Constituents

The C-H-O ternary diagram for the experimental simulator is shown in Figure 7. Appearing on the diagram are:

- . A point representing the simulator fuel composition, with coordinates derived from Formula (3).
- . A tie line connecting the fuel point with points representing the solid and gas phase products of combustion predicted by thermochemical equilibrium theory.
- . Points representing the experimental analyses of the gaseous and solid exhaust products, from Tables 4 and 5 respectively.

There is an imbalance in the experimental gas and solid analyses, reflected in the fact that these last two points cannot be connected with a tie line passing through the fuel point. This can be interpreted as arising from a deficiency of oxygen atoms in the two exhaust products, relative to the measured oxygen flow to the combustor.

It is nevertheless abundantly clear, from both Table 4 and the graphical evidence of Figure 7, that the reactions which took place in the experimental simulator fell well short of reaching thermochemical equilibrium. Comparison of Figures 6 and 7 also suggests, at least as indicated by the gas phase compositions, that the simulator efflux was further from equilibrium than that of the solid gas generator in Reference 4.

Notwithstanding the apparent imbalance in the measured oxygen content of the fuel and exhaust products, the total solid:gas mass fraction in the efflux can be estimated on the basis of measured carbon and hydrogen atom fractions. The procedure is set out in Appendix 2. The calculated ratio of 0.16:0.84 is not inconsistent with the result of the "dry" solid sampling experiment discussed in the previous Section, particularly in view of the likelihood of that result having been affected by loss of material during treatment of the captured sample. The above solid:gas mass ratio is about half the ratio estimated from Zaccardi and McGregor's results. This is consistent with the use of a less fuel-rich input mixture in the present experiments, and may also be due in part to the reaction in the simulator having been arrested further from thermochemical equilibrium.

9.3 Assessment of Results and Future Work

As has already been noted, the results of the present work are not directly comparable with those of Reference 4, because the mixture burned in the experimental simulator was significantly less fuel-rich than the solid propellant gas generator. Moreover, the above analyses of the results involved a measure of speculation, especially in the case of the Reference 4 data. Nevertheless, on the basis of these experiments some observations can be made which are pertinent to the level of simulation likely to be achievable with the approach adopted, and to directions for future work.

With simulator fuels chosen simply on the basis of elemental composition, not only does thermodynamic theory predict products of combustion which are different from those in the solid gas generator, but the relationship between thermodynamic theory and actual composition of the exhaust products also appears to be different for the two fuels. This is perhaps not surprising, since the reaction paths associated with production of the efflux constituents from the solid fuel grain must be different

from those associated with the simulator fuels. Further, the choice of simulator fuels can have a direct bearing on the tendency to form carbon, even though the fuel mixture may have C:H ratios which are identical to those of the solid propellant. Fuels which are of the benzene series (eg. toluene) have many times the tendency to form soot than do fuels from the olefin series (eg. ethylene)⁸, a characteristic not predicted by thermochemical equilibrium codes.

Differences in the physical processes preceding chemical reaction are also likely to be significant. In the case of the solid propellant, the fuel is intimately mixed with the oxidant, while in the simulator, the fuel must vaporise (if a liquid) and at least partially mix with the oxidant before combustion can take place. Combustion residence time is clearly important in both systems. For the solid gas generator, this is confirmed by observed variations in the efflux composition as the propellant grain was consumed and the combustion volume simultaneously increased⁴. The rate of reaction in liquid fueled gas generators, while generally fast for hydrocarbon fuels, is limited by availability of energy for mechanical mixing of the fuel and oxygen flows⁹. Clearly, residence time is a variable which must be investigated.

It will also be necessary to investigate the effect of varying fuel composition, in terms of both mixture proportions and constituent compounds. Experiments with more realistic fuel/oxygen ratios are required, perhaps with spatial redistribution of oxygen injection to increase the relative flame temperature in the upper pilot zone. The use of other feed fluids, such as benzene, methane, and/or carbon dioxide, may also be beneficial.

There is a need for more data on the efflux properties of solid hydrocarbon gas generators, to improve the basis for development of the simulator. This requires reliable sampling and analysis of both gaseous and solid phase products. Likewise, the exhaust sampling methods used in the present work need to be refined. For example, capturing gas samples from flush holes in the combustor walls yields results which are likely to be subject to errors arising from local non-uniformity of the combustion products. Methods need to be developed which ensure that the samples analysed are representative of the material which would fuel the secondary combustion process in ramburner experiments using the simulator. This includes solid particulate matter which must be analysed both chemically and physically for comparison with the real gas generator output. The possibility of chemical changes

occurring in the captured samples prior to analysis, either within the sampling system or during storage, cannot be discounted.

Although it may not provide a total analysis, a mass spectrometer would be invaluable for providing a real time gas analysis capability. It would assist to remove the uncertainty, when employing gas chromatography alone, due to the elapsed time between sampling and subsequent analysis. In addition, a real time instrument could possibly be configured to provide information on the elemental composition of the solid phase.

For simple hydrocarbon fuel formulations, relative masses of the solid and gaseous product phases can be calculated if accurate chemical analyses of both materials are available, and if the analyses can be shown to balance on the C-H-O ternary diagram. In these circumstances quantitative measurement of the relative rates of production of the different phases are arguably redundant. In view of the experimental difficulties associated with such measurements, and the consequent potential for error in the results, it would seem prudent to forgo further development of these techniques in favour of perfecting the methods for capturing and analysing truly representative samples to determine their chemical composition, as discussed above.

10. CONCLUSIONS

- (a) A relatively low pressure hydrocarbon gas generator simulator was developed, which burned a mixture of ethylene, toluene and oxygen. The simulator proved to be a reliable research tool which was both easy and safe to operate. Hardware refinements have been identified which would be desirable for future work.
- (b) The choice of fuels proved to represent a good balance between composition, availability and safety in handling.
- (c) The experimental simulator emitted a high temperature mixture of particulate and gaseous products which was qualitatively representative of current solid hydrocarbon gas generator design. The efflux was easily and cleanly disposed of in an afterburning ejector.

- (d) The quantitative determination of exhaust composition easily rivalled the simulator hardware development in terms of difficulty and time required to obtain meaningful results.
- (e) The exhaust composition differed significantly from that predicted by thermochemical equilibrium theory, as does the efflux from solid gas generators.
- (f) Comparison of the experimental results with available data from solid gas generator firings has provided guidance for further development of the simulator, in terms of geometry and fuel composition.
- (g) There is a need for more data on the efflux properties of solid hydrocarbon gas generators, to improve the basis for simulator development.
- (h) Techniques used in the present work for sampling and analysis of both gaseous and solid phase exhaust products need to be refined, as a prelude to further simulator development. A mass spectrometer would be advantageous.
- (i) Provided that accurate chemical analyses of the fuel mixture and representative exhaust samples are available, quantitative measurement of the relative rates of production of the solid and gaseous exhaust phases may not be essential.
- (j) This low pressure simulator has established the feasibility of the concept and indicated developments necessary to provide a useful simulator.

11. ACKNOWLEDGEMENT

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APPENDIX 1Mass Balance in Conjectured Solid Gas Generator Exhaust

The atom fractions of the elemental constituents in the fuel and two product phases are taken to be as follows:

	C	H	O	
Fuel	0.363	0.531	0.106	(from Formula (1))
Gas Phase	0.282	0.563	0.155	(from Table 3, measured data)
Solid Phase	0.540	0.460	-	(from Figure 6)

If n is the fraction of total fuel atoms which go to the gaseous phase and $(1 - n)$ the fraction of fuel atoms which go to the solid phase, then based on the above figures for carbon:

$$0.282n + 0.540(1 - n) = 0.363, \text{ giving } n = 0.686$$

A similar result can be calculated from the hydrogen and oxygen atom fractions in the above Table.

Taking $n = 0.69$ and introducing the respective atomic masses of carbon, hydrogen and oxygen:

$$\frac{\text{Mass of solid phase}}{\text{Mass of gas phase}} = \frac{0.31 (0.540 \times 12 + 0.460 \times 1)}{0.69 (0.282 \times 12 + 0.563 \times 1 + 0.155 \times 16)}$$

$$= \frac{0.33}{0.67}$$

APPENDIX 2Mass Balance in Experimental Simulator Exhaust

According to experimental measurements, the atom fractions of the elemental constituents in the fuel and two product phases are:

	C	H	O	
Fuel	0.352	0.506	0.142	(from Formula (3))
Gas Phase	0.327	0.536	0.137	(from Table 4, measured data)
Solid Phase	0.605	0.359	0.036	(from Table 5, ignoring nitrogen)

As discussed in Section 9.2.4 and illustrated in Figure 7, these figures contain an overall imbalance of C:H:O, particularly in respect of oxygen. The solid:gas mass fraction in the efflux may still be estimated using the figures for carbon and hydrogen.

As in Appendix 1, define n as the fraction of total fuel atoms which go to the gaseous phase, so that $(1 - n)$ is the fraction of fuel atoms which go to the solid phase.

Based on the above atom fractions for carbon:

$$0.327n + 0.605(1 - n) = 0.352, \text{ giving } n = 0.91$$

And for hydrogen:

$$0.536n + 0.359(1 - n) = 0.506, \text{ giving } n = 0.83$$

Taking the mean of these figures ($n = 0.87$) and introducing atomic masses:

$$\frac{\text{Mass of solid phase}}{\text{Mass of gaseous phase}} = \frac{0.13 (0.605 \times 12 + 0.359 \times 1 + 0.036 \times 16)}{0.87 (0.327 \times 12 + 0.536 \times 1 + 0.137 \times 16)}$$

$$= \frac{0.16}{0.84}$$

CONSTITUENT MOLE FRACTIONS (%)

	Solid Propellant	Formula (2) Mixture
Al ₂ O ₃	0.2	0.0
C	43.2	41.8
CH ₄	7.0	4.2
CO	11.8	16.3
CO ₂	1.0	0.3
FeCl ₂	0.3	0.0
HCl	3.8	0.0
HF	0.5	0.0
H ₂	26.0	35.9
H ₂ O	3.9	1.4
N ₂	2.2	0.0

GAS PROPERTIES

Temperature (K)	1285	1476
Molecular Mass	24.8	19.5
Gamma	1.14	1.17

TABLE 1

THEORETICAL PREDICTION OF EFFLUX

COLUMN	OVEN TEMP, °C	GASES
Silica Gel, 3mm dia x 5.5m	60	H ₂
		CO
		CH ₄
		C ₂ H ₆
		CO ₂
		C ₂ H ₄
		C ₂ H ₂
Molecular Sieve 5A, 3mm dia x 5.2m	30	O ₂
		N ₂
Porapak Q, 3mm dia x 1.5m	50	H ₂ O
	200	C ₆ H ₆
		C ₇ H ₈

TABLE 2

GAS CHROMATOGRAPHY DETAILS

CONSTITUENT MOLE FRACTIONS (%)

	Predicted	Measured
CH ₄	12.3	25.7
CO	21.4	28.5
CO ₂	1.7	9.7
C ₆ H ₆	0.0	4.1
FeCl ₂	0.5	0.5*
HCl	6.5	2.6
HF	1.2	1.2*
H ₂	45.9	22.6
H ₂ O	6.4	0.8
N ₂	3.8	2.5

* Not measured, but assumed to be the same as the theoretical value.

TABLE 3 ANALYSIS OF EXHAUST GAS FROM REFERENCE 4

CONSTITUENT MOLE FRACTIONS (%)

	Predicted	Measured
CH ₄	0.4	5.1
CO	36.0	38.6
CO ₂	0.0	2.4
C ₂ H ₂	0.0	2.7
C ₂ H ₄	0.0	2.7
C ₂ H ₆	0.0	0.1
C ₆ H ₆	0.0	3.3
C ₇ H ₈	0.0	3.8
H ₂	63.4	41.2
H ₂ O	0.2	0.0
N ₂	0.0	0.4
O ₂	0.0	0.0

TABLE 4 ANALYSIS OF EXHAUST GAS FROM PRESENT EXPERIMENT

Constituent	Measured Mass Fraction	Resultant Atom Fraction
C	0.863	0.596
H	0.043	0.354
N	0.024	0.014
O	0.068	0.035

TABLE 5 ANALYSIS OF SOLID SAMPLE FROM PRESENT EXPERIMENT

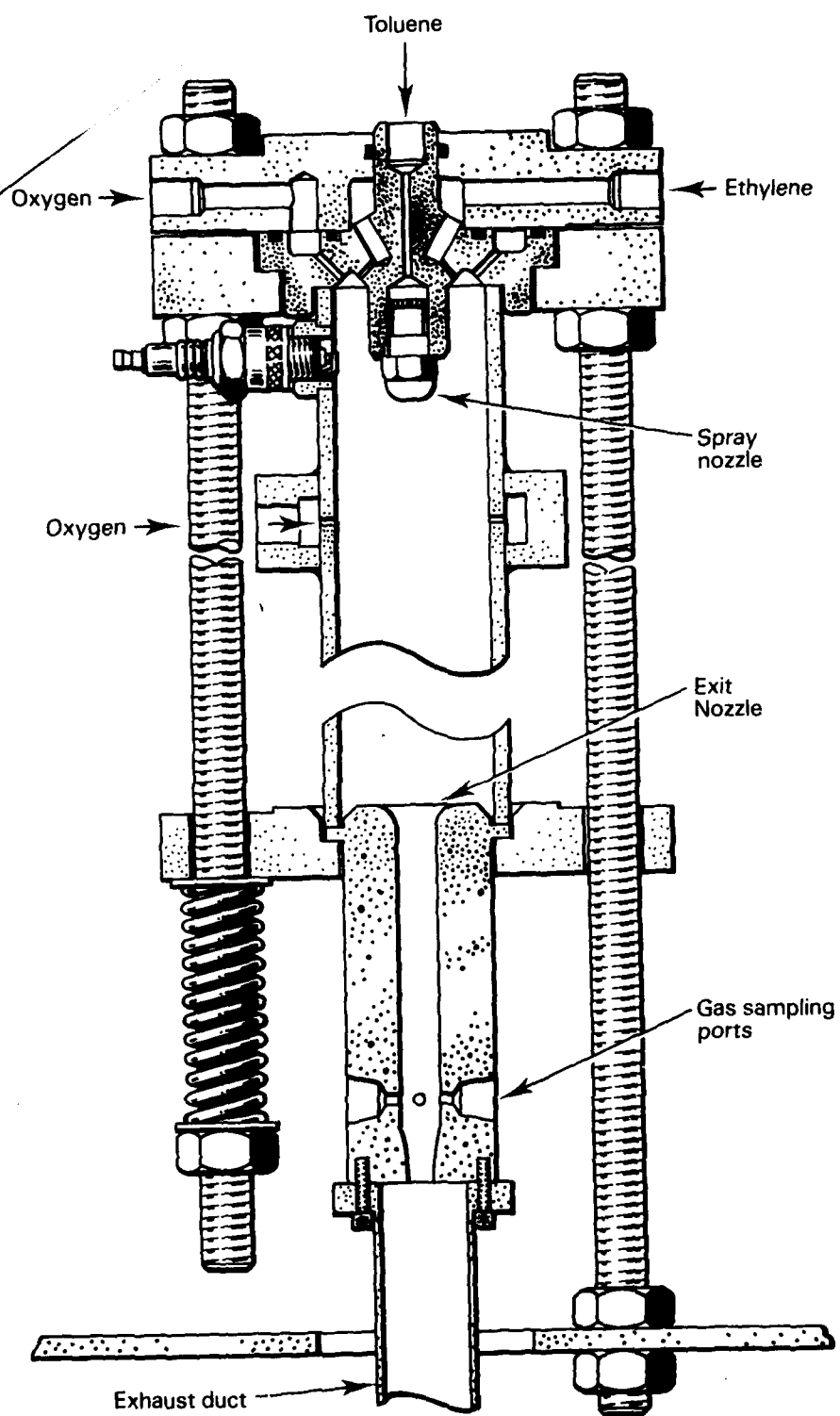
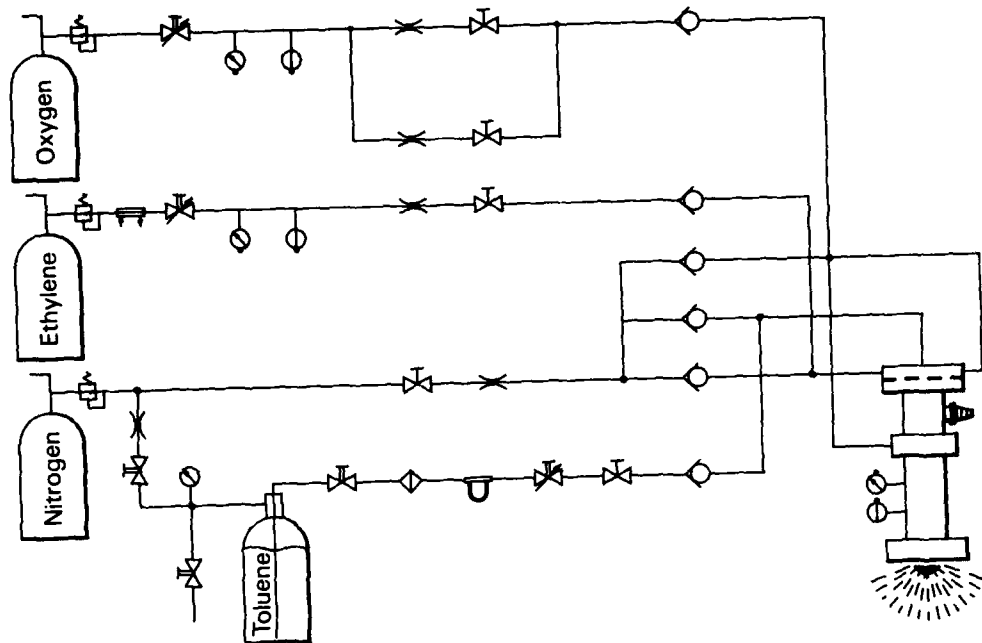


FIG. 1 EXPERIMENTAL COMBUSTOR



KEY













-  Pressure regulator
-  Manual control valve
-  Manual on/off valve
-  Pneumatic on/off valve
-  Check valve
-  Orifice
-  Filter
-  Mass flowmeter
-  Pressure gauge
-  Pressure transducer
-  Thermocouple
-  Heat exchanger

FIG. 2 FUEL SUPPLY SYSTEM

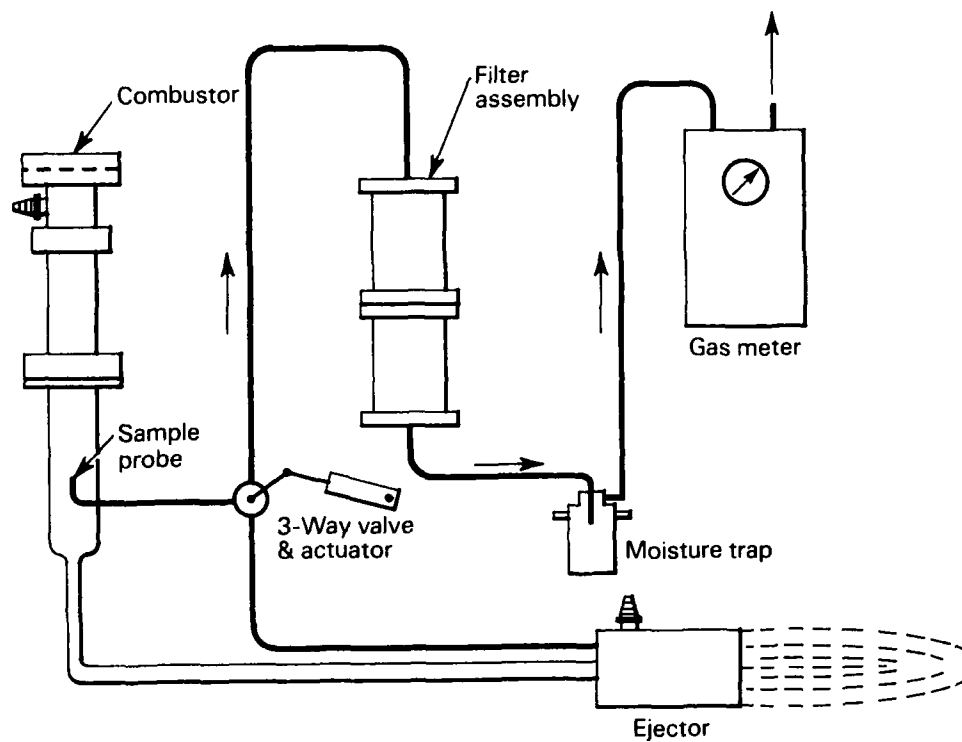


FIG. 3 EXHAUST DUCTING AND PARTICLE SAMPLING SYSTEM

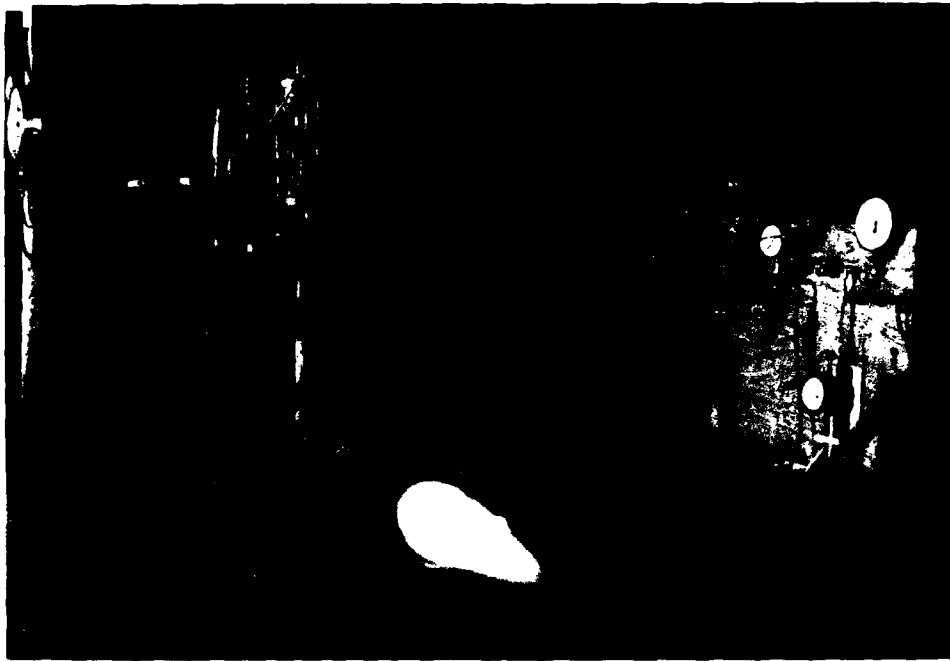


FIG. 4 PHOTOGRAPH OF RIG IN OPERATION

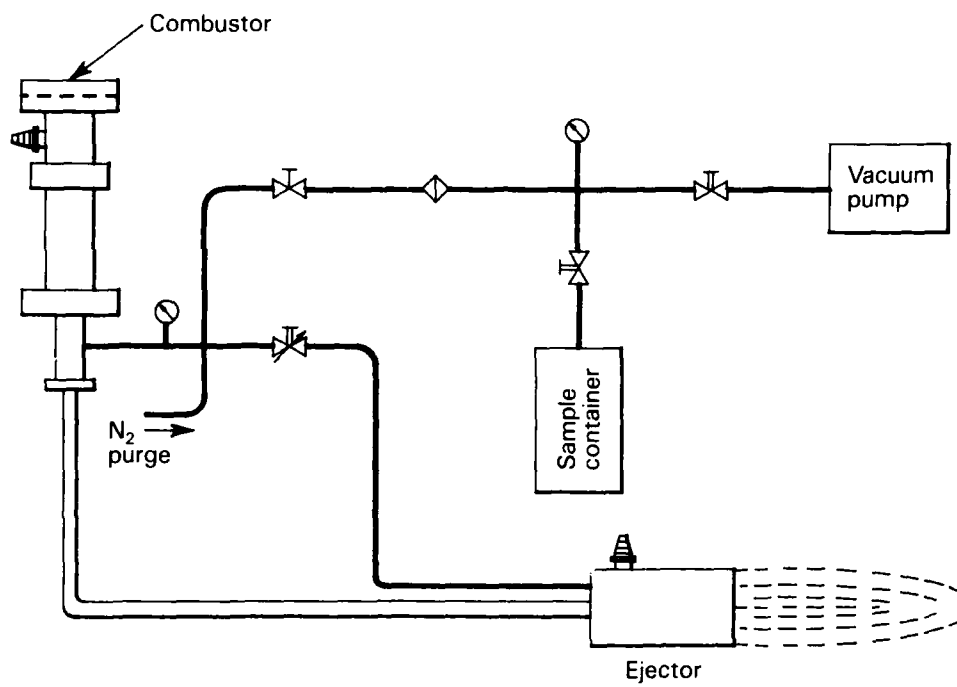
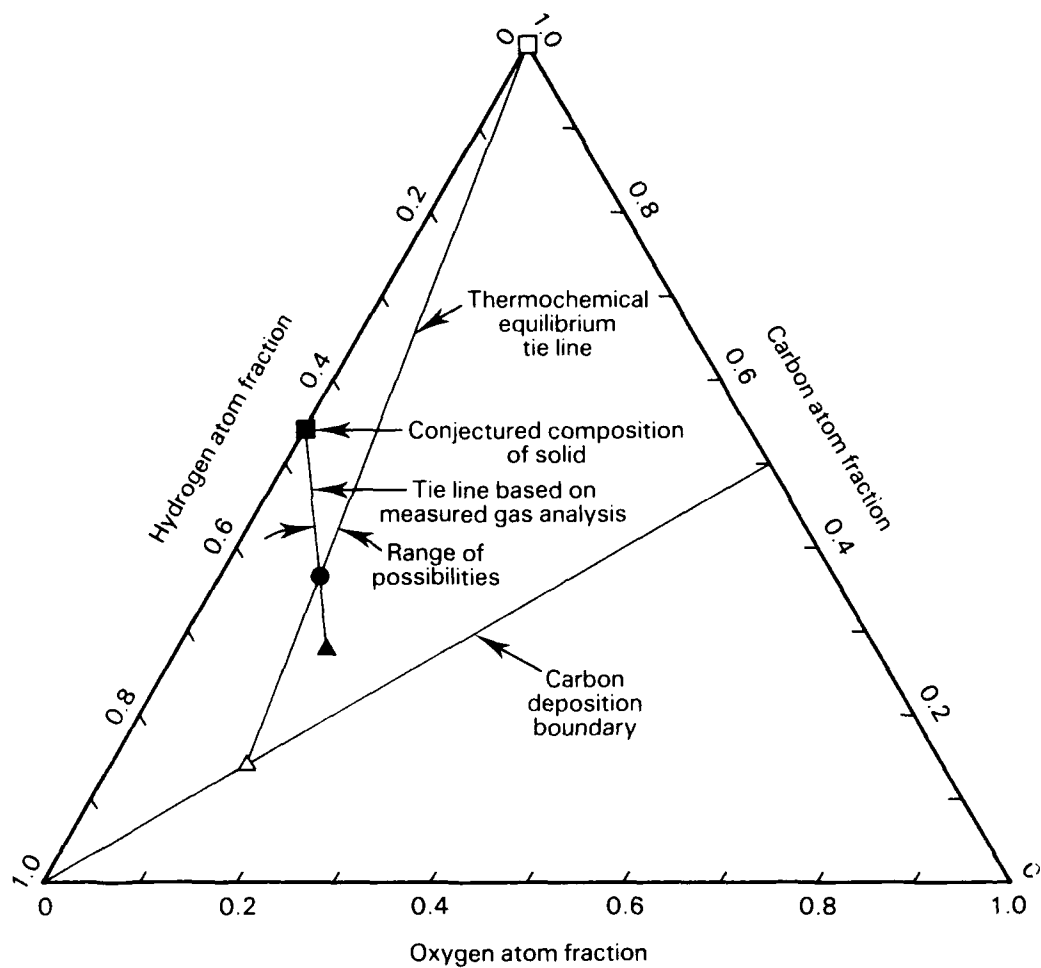


FIG. 5 EXHAUST GAS SAMPLING SYSTEM



- Fuel
- △ ▲ Reaction product – gas phase
- ■ Reaction product – solid phase
- ↑ Experiment
- Thermochemical equilibrium

FIG. 6 C-H-O TERNARY DIAGRAM – REFERENCE 4 RESULTS

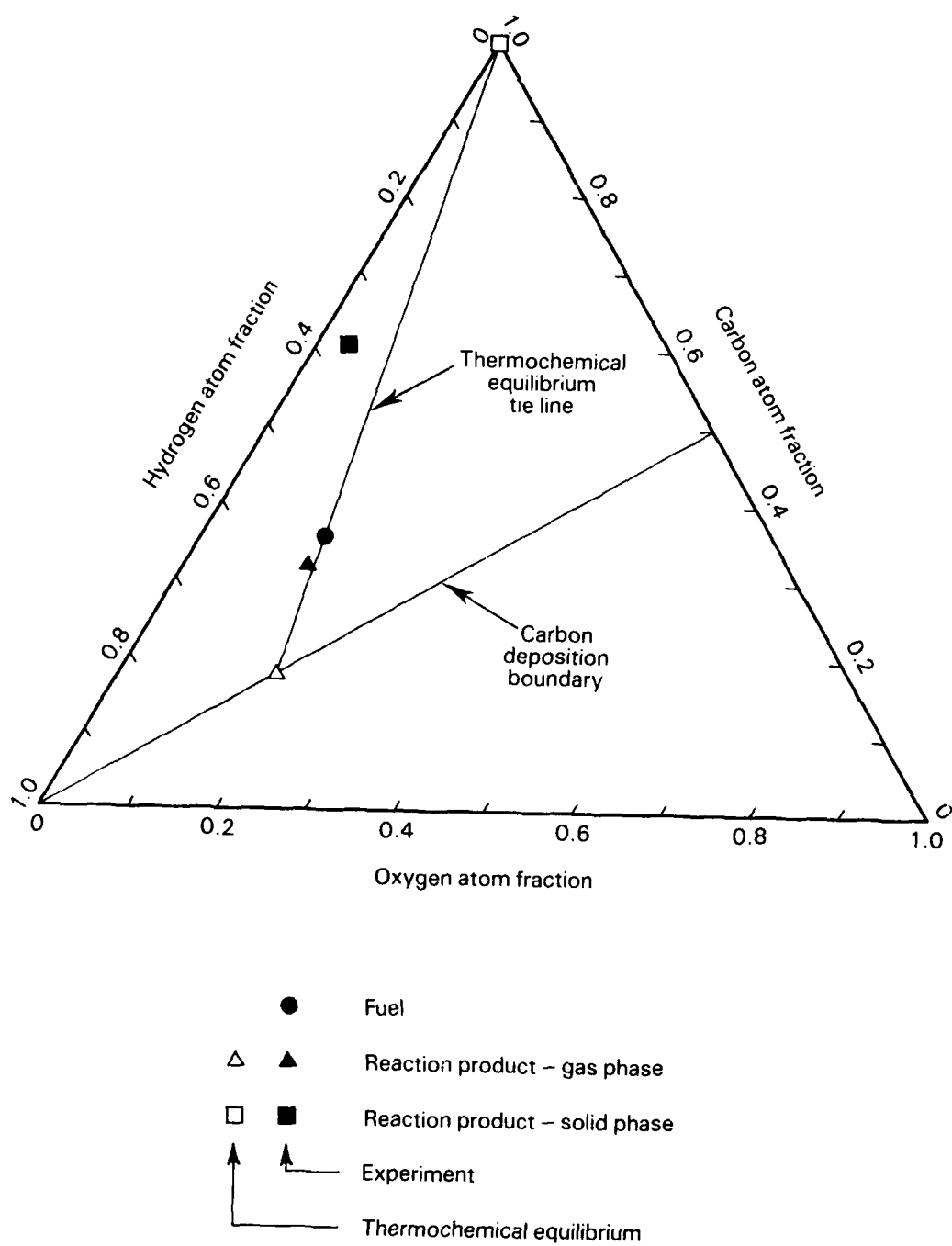


FIG. 7 C-H-O TERNARY DIAGRAM – PRESENT EXPERIMENT

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